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Vertical correlations and anti-correlations in multi-layered arrays of 2D quantum islands

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Recent breakthroughs in quantum wire- and quantum dot fabrication rely considerably on effects of spontaneous formation of ordered nanostructures [1,2]. Multi-sheet arrays of 3D or 2D islands are distinct from other types of nanostructures since the formation of such an array is governed by *both equilibrium ordering and kinetic-controlled ordering*. If the deposition of the first sheet of islands of material 2 on a material 1 is followed by a growth interruption, islands of an equilibrium periodic structure are formed [3,4]. If the islands are regrown by material 1, and material 2 is again deposited, a new growth mode occurs. For typical growth temperatures and growth rates, the structure of the buried islands of the first sheet does not change during the deposition of the second sheet. The second sheet of islands grows *in the strain field created by the buried islands of the first sheet.* And the structure of the second sheet reaches the equilibrium *under the constraint of the fixed structure of the buried islands of the first sheet.*

A remarkable feature of multi-sheet arrays of *3D islands* is that the buried islands in successive sheets are spatially correlated. At the surface, new islands were observed to be formed directly above buried islands [5–7]. The existing theory explaining well the correlation accounts the strain created by buried islands and shows that energetically preferred sites for nucleation of islands of the second sheet occur above buried islands [6,7]. In [6,7], buried islands were approximated as elastic point defects, and the crystal was treated as elastically isotropic medium.

In seeming contradiction to the above results, very recent experiments on multi-sheet arrays of 2D islands of CdSe in ZnSe matrix [8] unambiguously and surprisingly revealed vertical *anti-correlation* between islands in successive sheets. Surface islands are formed above the spacings in the sheet of buried islands.

Here we examine the energetics of multi-sheet arrays of 2D islands and seek the equilibrium configuration of the array of surface islands, under the constraint of a fixed array of buried islands. The two key inputs of our treatment which make it different from those of [6, 7], are as follows. *i)* We consider 2D islands of 1–2 monolayers height where the separation between successive sheets is comparable or even less than the lateral size of the islands, and we take into account their exact shape. *ii)* We take into account the elastic anisotropy of cubic crystals in question.

Since a single-sheet array of 2D islands can exist both as a 1D array of stripes and as a 2D array of compact islands (disks) [9], we address both possibilities. To extract essential physics, it suffices to examine a double-sheet array comprised of one sheet of buried islands and one sheet of surface islands. The total energy of the double-sheet array equals,

$$E_{\text{total}} = E_{\text{surf}} + E_{\text{boundaries}} + \Delta E^{(SS)}_{\text{elastic}} + E^{(SB)}_{\text{elastic}} . \qquad (1)$$

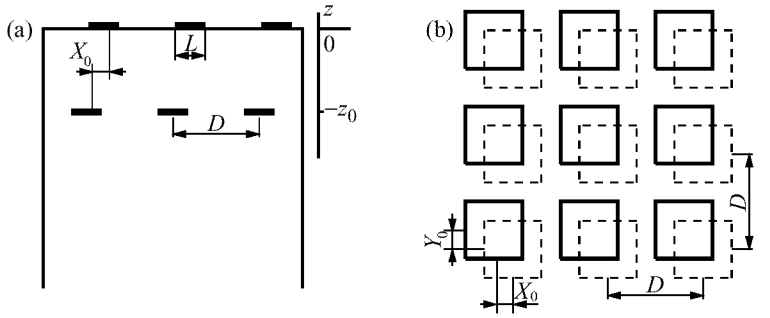

Fig 1. Geometry of double-sheet arrays of 2D islands. (a) Each sheet of islands forms a 1D array of stripes. The cross-section is shown. (b) Each sheet of islands forms a 2D array of square-shaped islands. The plan view is plotted. Buried islands are depicted by dashed lines, and solid lines are used for surface islands.

Here E_{surf} is the sum of the surface energies of surface islands and of uncovered parts of material 1, $E_{\text{boundaries}}$ is the energy of island boundaries, $\Delta E^{(SS)}_{\text{elastic}}$ is the elastic relaxation energy of surface (S) islands due to the discontinuity of the intrinsic surface stress tensor on island boundaries [3,9], $E^{(SB)}_{\text{elastic}}$ is the elastic energy of the interaction of surface islands (S) and of buried islands (B). We focus on the typical experimental situation of *an equal amount of the deposited material in each deposition cycle*. Then each sheet of islands alone tends to form the *same periodic structure* which corresponds to the minimum of the sum of the first three terms of Eq.(1). If the interaction between the two sheets is neglected, the surface array of islands *as a whole* can be subject to an arbitrary shift in the xy-plane. The strain due to buried islands has the same periodicity as the array of surface islands. Therefore the fourth term in Eq.(1) does not change the periodicity of the surface structure, and just defines its *relative position with respect to the array of buried islands* (Fig. 1). Since the interaction energy $E^{(SB)}_{\text{elastic}}$ is the only term in Eq.(1) which depends on the relative shift of the two arrays we focus only on this energy term as a function of the shift X_0 for 1D array of stripes (Fig. 1a) and of the shift (X_0, Y_0) for 2D array of compact islands (Fig. 1b).

To evaluate the strain due to buried islands, we refer to the strain due to point defects [10]. A point defect located at $\widetilde{\mathbf{r}}$ is represented by the superposition of three mutually perpendicular double forces (by an elastic dipole), and the effective body force density is $f_i(\mathbf{r}) = a_{ij}\nabla_j\delta(\mathbf{r} - \widetilde{\mathbf{r}})$. A monolayer-thick inclusion in the plane $z = \widetilde{z}$ with macroscopic lateral dimensions is a 2D array of point defects. It can be described by a 2D shape function $\Theta^B(\mathbf{r}_{\|})$ which equals 1 inside the inclusion, and 0 otherwise. The body force density associated with a 2D inclusion can be obtained by adding contributions of single point defects,

$$f_i(\mathbf{r}) = \frac{1}{A_0}\int d^2\widetilde{\mathbf{r}}_{\|}\, a_{ij}\nabla_j\left[\delta(\mathbf{r}_{\|} - \widetilde{\mathbf{r}}_{\|})\delta(z - \widetilde{z})\right]\Theta^B(\widetilde{\mathbf{r}}_{\|})\,, \tag{2}$$

where A_0 is unit cell area in the xy plane. Equation (2) is derived under the assumption of no mutual influence between the point defects comprising the inclusion. Generally speaking, the tensor a_{ij} characterizing the double force density is different for a single point defect and for a monolayer-thick inclusion. A substitutional impurity atom in

a zinc blend crystal of III–V or II–VI semiconductor has T_d site symmetry, and the corresponding tensor a_{ij} has the cubic symmetry. On the other hand, if the inclusion of equal substitutional impurity atoms is oriented in the (001) plane of the zinc blend crystal, has monolayer thickness and infinite lateral dimensions, *each atom of the inclusion has D_{2d} symmetry*. Therefore, the tensor a_{ij} characterizing a monolayer-thick buried island has a *uniaxial symmetry*.

The elastic properties of the surface islands are described by the difference of two-dimensional intrinsic surface stress tensors ($\Delta\tau_{\alpha\beta}$) of the two materials. The energy of the elastic interaction between a periodic array of buried islands and a similar periodic array of surface islands is obtained in the form of the sum over the reciprocal lattice vectors [11]

$$E_{\text{elastic}}^{(SB)} = \frac{h^B}{2A_0}\sum_{\mathbf{k}_\parallel}\left|\widetilde{\Theta}(\mathbf{k}_\parallel)\right|^2 \exp\left(i\mathbf{k}_\parallel\mathbf{R}_0\right)(\Delta\tau_{\alpha\beta})a_{lm}\nabla'_m \left[\nabla_\alpha\widetilde{G_{\beta l}}(\mathbf{k}_\parallel;z,z') + \nabla_\beta\widetilde{G_{\alpha l}}(\mathbf{k}_\parallel;z,z')\right]\Big|_{\substack{z=0\\z'=-z_0}}, \tag{3}$$

where $\mathbf{R}_0$ is the relative sheet of the two arrays, h^B is the thickness of the buried islands, $\nabla_x \equiv ik_x$, $\nabla'_x \equiv -ik_x$, $\nabla_y \equiv ik_y$, $\nabla'_y \equiv -ik_y$, α, β=1,2, l, m=1,2,3. We treat the crystal as elastically anisotropic cubic medium and use the static Green's tensor $\widetilde{G_{il}}(\mathbf{k}_\parallel;z,z')$ from [12]. The dependence of $E_{\text{elastic}}^{(SB)}$ on the separation between the two sheets is determined by the behavior of $\widetilde{G_{il}}(\mathbf{k}_\parallel;z,z')$ as a function of z_0. $\widetilde{G_{il}}(\mathbf{k}_\parallel;z,z')$ is a linear combination of three exponentials, $\exp(-\alpha_s kz_0)$, where three attenuation coefficients α_s are functions of the direction $\mathbf{k}_\parallel$ in the surface plane [12]. The key point is that, in a cubic crystal with a negative parameter of elastic anisotropy, $\Delta = (c_{11} - c_{12} - 2c_{44})/c_{44} < 0$, which is the fact for all III–V and II–VI cubic semiconductors, two of the three α_s are complex conjugate. Complex attenuation coefficients α imply that *the static analogues of Rayleigh waves exhibit not purely an exponential decay, but an oscillatory one*. This phenomenon is known for surface acoustic waves which are *generalized Rayleigh waves* in elastically anisotropic crystals [13]. Complex attenuation coefficients lead to the conclusion that the elastic interaction between successive sheets of islands exhibits an oscillatory decay with the separation between sheets.

The interaction energy (3) has been evaluated for double-sheet arrays of stripes and for double-sheet arrays of square islands. For the separation between the two sheets, $z_0 \leq 0.5D_0$ where D_0 is the lateral period, the difference between the value of $E_{\text{elastic}}^{(SB)}$ for the most favorable relative arrangement, and the one for the most unfavorable arrangement, is of the order of 0.1 meV/Å^2. This is the same order of magnitude as a typical energy of a single sheet of surface islands [9]. *This comparison confirms that the elastic interaction between the two sheets of islands can indeed result in vertical correlation or anti-correlation between the two sheets.*

The phase diagram of Fig. 2b shows that the favorable relative arrangement of the two sheets of islands *alternates from vertical correlation to anti-correlation*, some intermediate arrangements being possible for small spacing z_0. The separation corresponding to the *transition* from correlation to anti-correlation *depends dramatically on the anisotropy parameter $P^B \equiv a_{zz}/a_{xx}$ of the double force density characterizing buried islands*. Our results are in agreement with existing experimental data on

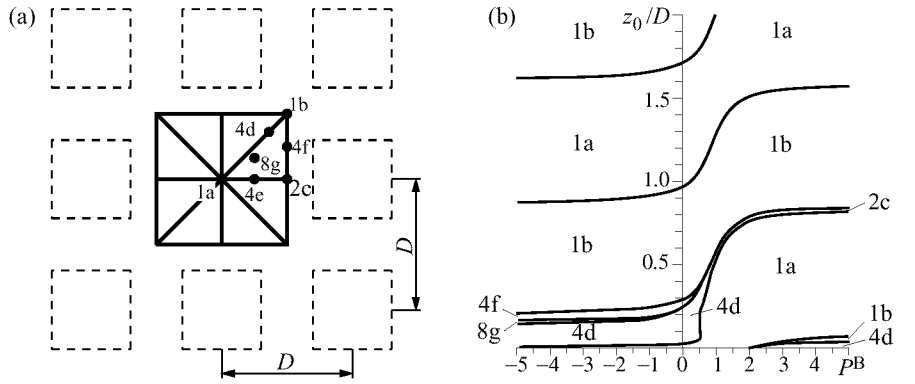

Fig 2. The phase diagram of a double-sheet array of square-shaped islands. (a) The relative shift (X_0, Y_0) is defined by the projection of the center of a surface island onto the superlattice formed by the buried islands. Seven types of the symmetry are labeled according to [14]. (b) The phase diagram for the surface coverage 0.35. P^B is the anisotropy of the double force density of buried islands, z_0 is the separation between the two sheets of islands, and D is the period.

anti-correlations in multi-sheet arrays of 2D islands [8].

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